# VERIFICATION OF TRANSLATION

Osaka-shi. Osaka 532-0003 JAPAN, hereby declare that I am conversant with the Japanese and English languages and that I am the translator of the documents attached and certify that to the best of my knowledge and belief the following is a true and correct English translation of the Japanese Patent Application No. 2002-214446 in the name of KANEKA CORPORATION.

Dated this 14 thday of September, 2006

Yasuo Yasutomi

SEP ? 6 2006

Japanesa Catent Application No. 2002-214446 filed on July 23, 2002

1

[Document Name] Patent Application [File Number] OSK-4804 [Filing Date] July 23, Heisei 14 (2002) [To] Commissioner, Patent Office [IPC] C08L 77/00 C08K 3/00 [Inventor] [Address or Residence] 2-54, Koyodai 1-chome, Kawanishi-shi, HYOGO "[Name] SUZUKI Noriyuki [Inventor] [Address or Residence] 55-1-103, Kuchitanihigashi 1-chome, Takarazuka-shi, HYOGO [Name] HARA Kazuhiro [Inventor] [Address or Residence] 5-35-509, Torikainishi, 5-chome, Settsu-shi, OSAKA [Name] ONO Yoshitaka [Applicant] [Identification Number] 000000941 [Name] KANEKA CORPORATION [Representative] TAKEDA Masatoshi [Indication of Fee] [Number of Deposit Ledger] 005027 [The Amount of Payment] 21000 yen [List of Attached Documents] [Document Name] Specification 1

Abstract

Needed

[Document Name]

[Necessity of Proof]

[DOCUMENT NAME] Specification
[TITLE OF THE INVENTON] POLYAMIDE RESIN COMPOSITON AND PROCESS
FOR PRODUCING THE SAME

- 5 [SCOPE OF CLAIUMS FOR PATENT]
  [Claim 1] A polyamide resin composition comprising polyamide
  resin and swelling mica treated with a polyether compound,
  wherein the polyether compound is represented by general
  formula (1):
- 10 [Chemical formula 1]

20

(wherein -A- represents -O-, -S-, -SO-, -SO<sub>2</sub>-, -CO-, an alkylene group having 1 to 20 carbon atoms, or alkylidene group having 6 to 20 carbon atoms; and  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  each represent a hydrogen atom, a halogen atom, or a monovalent hydrocarbon group having 1 to 5 carbon atoms, and the Rs may be the same or different).

[Claim 2] The polyamide resin composition of Claim 1, wherein 25 the polyether compound is represented by general formula (2): [Chemical formula 2]

$$R^{1} \leftarrow OR^{9} \rightarrow O \rightarrow R^{1} \rightarrow R^{5} \rightarrow R^{6} \rightarrow O \rightarrow R^{10}O \rightarrow R^{12} \qquad (2)$$

$$R^{3} \rightarrow R^{4} \rightarrow R^{7} \rightarrow R^{8}$$

(wherein -A- represents -O-, -S-, -SO-, -SO<sub>2</sub>-, -CO-, an alkylene group having 1 to 20 carbon atoms, or alkylidene group having

6 to 20 carbon atoms; and  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  each represent a hydrogen atom, a halogen atom, or a monovalent hydrocarbon group having 1 to 5 carbon atoms;  $R^9$  and  $R^{10}$  each represent a divalent hydrocarbon group having 1 to 5 carbon atoms;  $R^{11}$  and  $R^{12}$  each represent a hydrogen atom or a monovalent hydrocarbon group having 1 to 20 carbon atoms; and the Rs may be the same or different; m and n each represent the number of oxyalkylene repeating units; and  $2 \le m+n \le 50$ ).

5

15

[Claim 3] The polyamide resin composition of Claim 1 or 2,
10 prepared by mixing said resin and swelling mica treated with
a polyether compound.

[Claim 4] The polyamide resin composition of Claim 1, 2, or 3, wherein the ratio of the swelling mica having an equivalent circular diameter [D] of 3000 Å or less in the composition is 20% or more.

[Claim 5] The polyamide resin composition of Claim 1, 2, 3, or 4, wherein the average of the equivalent circular diameter [D] of the swelling mica in the polyamide resin composition is 5000 Å or less.

[Claim 6] The thermoplastic resin composition of Claim 1, 2, 3, 4 or 5, wherein the average layer thickness of the swelling mica in the polyamide resin composition is 500 Å or less.

[Claim 7] The polyamide resin composition of Claim 1, 2, 3, 4, 5 or 6, wherein the maximum layer thickness of the swelling mica in the polyamide resin composition is 2000 Å or less.

[Claim 8] The polyamide resin composition of Claim 1, 2, 3,

[Claim 8] The polyamide resin composition of Claim 1, 2, 3, 4, 5, 6 or 7, wherein the number [N] of particles per unit weight ratio of the swelling mica in the polyamide resin composition is 30 or more.

30 [Claim 9] The polyamide resin composition of Claim 1, 2, 3, 4, 5, 6, 7 or 8, wherein the average aspect ratio (layer length/layer thickness) of the swelling mica in the polyamide resin composition is 10 to 300.

[Claim 10] A method for making a polyamide resin composition, 35 comprising melt-mixing the polyamide resin composition set forth in claim 1.

[Detailed Description of the Invention]
[0001]

5 [Technical Field to which the Invention Pertains]

The present invention relates to polyamide resin compositions containing polyamide resins and swelling mica treated with particular polyether compounds.

[0002]

10 [Background Art]

15

20

25

30

35

Polyamide resins have high resistance to heat, chemicals, and weathering and exhibits, for example, excellent mechanical and electrical properties. Polyamide resins are thus used in various industrial applications, such as injection molding materials, fibers, and films. However, polyamide resins are highly crystalline. When they are formed into a thin product, a complex-shaped product having nonuniform thickness, or the like by injection molding, the product suffers from problems such as warpage resulting in deformation of the product. In order to overcome such problems, in general, incorporation of various inorganic particles has been attempted for improvement. However, this approach also has a problem such as poor surface appearance of the product or orientation of fibrous inorganic substances resulting in anisotropy and thus deformation of the product.

[0003]

Such problems of inorganic particles are presumably caused by insufficient dispersibility of inorganic particles or excessive size of dispersed particles. A technique for finely dispersing inorganic particles has been desired.

[0004]

Japanese Unexamined Patent Application Publication Nos. 62-74957, 2-69562, 6-80873, 6-228435, 11-349811, 6-248176, 8-283567, 9-241505, and 2001-2913 disclose polyamide resin compositions containing swelling clay compounds, i.e.,

inorganic particles, finely dispersed in polyamide resins. In these techniques, swelling clay compounds, such as montmorillonite, are added during the polymerization for polyamide resins. However, clay compounds increase the melt viscosity of the polymer, thereby causing agitation failure during the polymerization. Thus, the amounts of clay compounds that can be used in the polymerization process are as small as approximately several percent, and this has limited the scope of the product design. Moreover, in order to compound auxiliary materials such as a flame retarder and a stabilizer, additional steps are necessary or the process becomes complicated. An improvement as to these points is also desired.

[0005]

10

15

20

25

30

Another problem of the above-described techniques is the use of organic ammonium salts as a surface treatment agent for achieving homogeneous, fine dispersion of swelling clay compounds. Organic ammonium salts retained at a temperature for processing polyamide resins for a long time undergo deterioration, thereby degrading the properties such as mechanical properties or toughness. This problem must also be overcome.

[0006]

Japanese Unexamined Patent Application Publication Nos. 8-319417, 2000-212432, 2000-290500, and 2001-302845 and WO 97/11998 disclose polyamide resin compositions containing swelling clay compounds finely dispersed in polyamide resins by extrusion. However, in these inventions, dispersion is insufficient, and thus the properties are not sufficiently improved. Moreover, the use of organic ammonium salts as a surface treatment agent for clay compounds may lead to deterioration during the processing and may degrade the properties such as mechanical properties or toughness. Among the above-described inventions, the invention set forth in Japanese Unexamined Patent Application Publication No.

35 2000-212432 requiring end-capped nylon and the invention set

forth in WO 97/11998 requiring organic ammonium salts having bicyclo rings are not suitable for industrial applications. [0007]

Japanese Unexamined Patent Application Publication No. 9-118518 discloses a technique for facilitating fine dispersion by rendering layers of swelling clay compounds readily cleavable. In this technique, a polymeric compound (intercalant polymer), such as polyvinylpyrrolidone, is intercalated between layers of sheet silicate to prepare an intercalation compound. However, although this invention provides the intercalation compound, it does not provide a technique of cleaving the intercalation compound and finely dispersing the cleaved intercalation compound into a polyamide resin. It has been difficult to finely disperse a swelling clay compound into a polyamide resin.

[8000]

5

10

15

20

25

30

Japanese Unexamined Patent Application Publication Nos. 10-259016 and 10-310420 disclose that, in order to cleave a layered swelling clay compound so that it can be finely dispersed in a thermoplastic resin, it is particularly effective to treat the swelling clay compound with a water-soluble compound to convert the compound into an intercalation clay compound. According to this technique, the elastic modulus and heat resistance can be improved without impairing surface appearance. However, further improvements on various properties and on warpage that occurs during injection molding are strongly desired.

[0009]

As is apparent from the above, there has been no technique of homogeneously and finely dispersing a swelling clay compound in a polyamide resin by a simple process, such as melt-kneading, to obtain a polyamide resin composition having excellent properties.

[0010]

35 [Problem to be solved by the Invention]

An object of the present invention is to overcome the above-described problems experienced in the conventional art by providing a polyamide resin composition having high dimensional stability, satisfactory mechanical properties, and high heat resistance.

[0011]

5

10

15

20

35

[Means for solving problem]

The present inventors have conducted extensive investigations to achieve these objects and completed the invention by providing a polyamide resin composition with superior properties, in which swelling mica treated with a particular polyether compound is finely and homogeneously dispersed in a polyamide resin by extrusion.

[0012]

In particular, the first aspect of the present invention provides a polyamide resin composition comprising polyamide resin and swelling mica treated with a polyether compound, wherein the polyether compound is represented by general formula (1):

[0013]

[Chemical formula 3]

30 [0014]

(wherein -A- represents -O-, -S-, -SO-, -SO<sub>2</sub>-, -CO-, an alkylene group having 1 to 20 carbon atoms, or alkylidene group having 6 to 20 carbon atoms; and  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  each represent a hydrogen atom, a halogen atom, or a monovalent hydrocarbon group having 1 to 5 carbon atoms, and the Rs may

be the same or different).

[0015]

In a preferred embodiment of the above polyamide resin composition, the polyether compound is represented by general formula (2):

[0016]

[Chemical formula 4]

$$R^{1} \leftarrow OR^{9} \rightarrow O \rightarrow R^{1} \rightarrow R^{2} \rightarrow R^{5} \rightarrow R^{6} \rightarrow O \leftarrow R^{10}O \rightarrow R^{12} \qquad (2)$$

$$R^{3} \rightarrow R^{4} \rightarrow R^{7} \rightarrow R^{8} \rightarrow R^{10}O \rightarrow R^{12} \qquad (2)$$

15 [0017]

5

20

25

30

35

(wherein -A- represents -O-, -S-, -SO-, -SO<sub>2</sub>-, -CO-, an alkylene group having 1 to 20 carbon atoms, or alkylidene group having 6 to 20 carbon atoms; and  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  each represent a hydrogen atom, a halogen atom, or a monovalent hydrocarbon group having 1 to 5 carbon atoms;  $R^9$  and  $R^{10}$  each represent a divalent hydrocarbon group having 1 to 5 carbon atoms;  $R^{11}$  and  $R^{12}$  each represent a hydrogen atom or a monovalent hydrocarbon group having 1 to 20 carbon atoms; and the Rs may be the same or different; m and n each represent the number of oxyalkylene repeating units; and  $2 \le m+n \le 50$ ).

[0018]

In a more preferred embodiment of the above polyamide resin composition, the polyamide resin composition is prepared by mixing said resin and swelling mica treated with a polyether compound.

[0019]

In a yet more preferred embodiment of the polyamide resin composition, the ratio of the swelling mica having an equivalent circular diameter [D] of 3000 Å or less in the composition is 20% or more.

[0020]

In a paticularly preferred embodiment of the above polyamide resin composition, the average of the equivalent circular diameter [D] of the swelling mica in the polyamide resin composition is 5000 Å or less.

[0021]

In another preferred embodiment of the above polyamide resin composition, the average layer thickness of the swelling mica in the polyamide resin composition is 500 Å or less.

10 [0022]

5

25

In another preferred embodiment of the polyamide resin composition, the maximum layer thickness of the swelling mica in the polyamide resin composition is 2000 Å or less.

[0023]

In another preferred embodiment of the polyamide resin composition, the number [N] of particles per unit weight ratio of the swelling mica in the polyamide resin composition is 30 or more.

[0024]

In another preferred embodiment of the polyamide resin composition, the average aspect ratio (layer length/layer thickness) of the swelling mica in the polyamide resin composition is 10 to 300.

[0025]

The second aspect of the present invention also provides a method for making a polyamide resin composition, comprising melt-mixing the polyamide resin composition set forth above.

[0026]

[Mode for Carrying Out the Invention]

In the present invention, polyamide resins used are polymers that contain amide bonds (-NHCO-) in the main chains and melt by heating. Examples of such polyamide resins include polycaproamide (nylon 6), polytetramethylene adipamide (nylon 46), polyhexamethylene adipamide (nylon 66),

35 polyhexamethylene sebacamide (nylon 610), polyhexamethylene

dodecamide (nylon 612), polyundecamethylene adipamide (nylon 116), polyundecanamide (nylon 11), polydodecaneamide (nylon 12), polytrimethylhexamethylene terephthalamide (nylon TMHT), polyhexamethylene isophthalamide (nylon 6I),

polyhexamethylene terephthal/isophthalamide (nylon 6T/6I), polybis(4-aminocyclohexyl)methane dodecamide (nylon PACM12), polybis(3-methyl-4-aminocyclohexyl)methane dodecamide (nylondimethyl PACM12), polymetaxylylene adipamide (nylon MXD6), polyundecamethylene terephthalamide (nylon 11T),

MXD6), polyundecamethylene terephthalamide (nylon 11T), polyundecamethylene hexahydroterephthalamide (nylon 11T(H)), and copolymers and mixtures of these polyamides. Among these, nylon 6, nylon 46, nylon 66, nylon 11, nylon 12, and copolymers and mixtures of these polyamides are preferred. Aromatic polyamide resins may be used. The molecular weights of these polyamide resins are not particularly limited. In general, polyamide resins having relative viscosities of 0.5 to 5.0 measured in conc.  $H_2SO_4$  at 25°C are preferred. These polyamide resins may be used alone. Alternatively, two or more of these polyamide resins having different compositions or components and/or different relative viscosities may be used. Among these, Nylon 6, nylon 66, nylon 46, and MXD nylon are more preferable from the standpoints of strength, modulus of elasticity, cost, and the like.

[0027]

10

15

20

25 The swelling fluorinated mica used in the present invention may be produced by heating a mixture of talc and a silicofluoride or fluoride of sodium and/or lithium. A specific production method is disclosed in Japanese Unexamined Patent Application Publication No. 2-149415. In this method, talc is subjected to intercalation of a sodium ion and/or a lithium ion to obtain swelling mica. In particular, talc is mixed with a silicofluoride and/or a fluoride, and the resulting mixture is treated at approximately 700°C to 1,200°C to obtain swelling mica. In the present invention, swelling fluorinated mica produced by this method is particularly preferable. In

order to obtain swelling mica, sodium or lithium must be the metal contained in the silicofluoride or the fluoride. These may be used alone or in combination. The total content or the content of the silicofluoride and/or the fluoride mixed with talc is preferably 10 to 35 percent by weight of the entirety of the mixture. If the content is out of the range, yield of the swelling mica may be decreased. The swelling mica produced by the above-described method has a structure represented by general formula (3) below:

10  $\alpha$  (MF)· $\beta$  (aMgF<sub>2</sub>.bMgO)· $\gamma$ SiO<sub>2</sub> (3) (wherein M represents sodium or lithium;  $\alpha$ ,  $\beta$ ,  $\gamma$ , a, and b each represent a coefficient;  $0.1 \le \alpha \le 2$ ;  $2 \le \beta \le 3.5$ ;  $3 \le \gamma \le 4$ ;  $0 \le a \le 1$ ;  $0 \le b \le 1$ ; and a+b=1).

Alternatively, it is possible to add a small amount of alumina  $(Al_2O_3)$  during the process of producing the swelling mica used in the present invention so as to control the swelling property of the resulting swelling mica.

[0028]

5

15

20

25

30

35

These are substances having such properies that swells in water, polar solvents miscible with water at arbitrary ratios, and mixed solvents containing water and any of these polar solvents. In this invention, "swelling property" refers to the property of mica to absorb polar molecules between the layers, thereby increasing the interlayer distance or to extensively swell, thus leading to cleaving. Examples of the swelling mica include lithium taeniolite, sodium taeniolite, lithium tetrasilicic mica, and sodium tetrasilicic mica, or substitution products thereof, derivatives thereof, or mixtures thereof. The swelling mica in an initial aggregation state, i.e., before swelling, has a basal-plane spacing of approximately 10 to 17 Å and an average particle diameter of approximately 1,000 to 1,000,000 Å.

[0029]

The polyether compound used in the present invention includes a structure represented by general formula (1) in the

side chain and/or the main chain of a polyoxyalkylene compound such as polyoxyethylene or a polyoxyethylene-polyoxypropylene copolymer:

[0030]

5 [Chemical formula 5]

[0031]

(wherein -A- represents -O-, -S-, -SO-, -SO<sub>2</sub>-, -CO-, an alkylene group having 1 to 20 carbon atoms, or an alkylidene group having 6 to 20 carbon atoms; and  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  each represent a hydrogen atom, a halogen atom, or a monovalent hydrocarbon group having 1 to 5 carbon atoms, and they may be the same or different).

20 [0032]

1.5

25

Among the above-described polyether compounds, those having a structure represented by general formula (2) below are preferred from the standpoints of thermal stability, dispersibility of the lamellar substance, and ready availability:

[0033]

[Chemical formula 6]

$$R^{11} \leftarrow OR^{9} \rightarrow R^{3} \qquad R^{4} \qquad R^{7} \qquad R^{8} \qquad O \leftarrow R^{10}O \rightarrow R^{12} \qquad (2)$$

[0034]

35 (wherein -A-represents -O-, -S-, -SO-, -SO<sub>2</sub>-, -CO-, an alkylene

group having 1 to 20 carbon atoms, or an alkylidene group having 6 to 20 carbon atoms; and  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ ,  $R^6$ ,  $R^7$ , and  $R^8$  each represent a hydrogen atom, a halogen atom, or a monovalent hydrocarbon group having 1 to 5 carbon atoms;  $R^9$  and  $R^{10}$  each represent a divalent hydrocarbon group having 1 to 5 carbon atoms;  $R^{11}$  and  $R^{12}$  each represent a hydrogen atom or a monovalent hydrocarbon group having 1 to 20 carbon atoms; and the Rs may be the same or different; m and n each represent the number of oxyalkylene repeating units; and  $2 \le m+n \le 50$ ).

10 [0035]

15

20

25

30

35

The above-described polyether compounds may contain a functinal group or groups. The functinal group may be any one provided that it does not adversely affect the polyamide resin or swelling mica. Examples of the substituents include saturated or unsaturated monovalent or multivalent aliphatic hydrocarbon groups; groups containing ester bonds; an epoxy group; an amino group; a carboxyl group; carbonyl-terminated groups; an amide group; a mercapto group; groups containing sulfonyl bonds; groups containing sulfinyl bonds; a nitro group; a nitroso group; a nitrile group; halogen atoms; and a hydroxyl group. The polyether compounds may be substituted with one of these or two or more of these.

[0036]

The content of the substituent in the polyether compound is not particularly limited as long as the polyether compound is soluble in water or a polar solvent containing water. In particular, the solubility of the polyether compound is preferably 1 g or more, more preferably 2 g or more, yet more preferably 5 g or more, still more preferably 10 g or more, and most preferably 20 g or more in 100 g of water at room temperature.

[0037]

Examples of the polar solvent include alcohols such as methanol, ethanol, and isopropanol; glycols such as ethylene glycol, propylene glycol, and 1,4-butanediol; ketones such as

acetone and methyl ethyl ketone; ethers such as diethyl ether and tetrahydrofuran; amide compounds such as

N,N-dimethylformamide and N,N-dimethylacetamide; and other solvents such as pyridine; dimethylsulfoxide; and

N-methylpyrrolidone. Carbonic acid diesters such as dimethyl carbonate and diethyl carbonate may also be used. These polar solvents may be used alone or in combination.

[0038]

5

10

15

20

25

30

The amount of the polyether compound used can be prepared to enhance the affinity between the swelling mica and the polyamide resin and to sufficiently improve the dispersibility of the swelling mica in the polyamide resin composition. necessary, two or more polyether compounds having different functional groups may be used simultaneously. Thus, the amount of the polyether compound to be formulated cannot be limited by specific numerical values; however, the lower limit of the amount of the polyether compound to 100 parts by weight of the swelling mica is preferably 1 part by weight, more preferably 2 parts by weight, and most preferably 5 parts by weight. If the lower limit of the amount of the polyether compound is below 1 part by weight, fine dispersion of the swelling mica tends to be insufficient. The upper limit is not particularly limited, but it is no need to use more than 200 parts by weight to 100 parts by weight of the swelling mica because fine dispersion of the swelling mica also tends to be insufficient when the amount exceeds 200 parts by weight.

[0039]

In the present invention, the process for treating the swelling mica with the polyether compound is not particularly limited. For example, the method described below may be employed.

First, swelling mica and a dispersion medium are mixed. Here, the dispersion medium is either water or a polar solvent containing water.

35 [0040]

The method for mixing the swelling mica and the dispersion medium is not particularly limited. For example, the mixing may be performed using a conventional wet mixer. Examples of the wet mixer include high performance mixers having mixing blades rotating at high speeds; wet mills for wet-milling samples in the gaps between rotors and stators at high shear rate; mechanical pulverizers for wet processes using hard media; impact pulverizers for wet processes, in which samples are collided at high speeds using jet nozzles and the like; and 10 ultrasonic pulverizers for wet processes using ultrasonic waves. In order to achieve more efficient mixing, the number of revolutions for mixing may be increased to 1,000 rpm or more, preferably 1,500 rpm or more, and more preferably 2,000 rpm or more. Alternatively, the shear rate may be increased to 500 15 (1/s) or more, preferably 1,000 (1/s) or more, and more preferably 1,500 (1/s) or more. The upper limit of the number of revolutions is approximately 25,000 rpm, and the upper limit of the shear rate is approximately 500,000 (1/s). Since mixing or application of shear beyond the upper limits does not improve 20 the mixing efficiency, there is no need to conduct mixing beyond the upper limits. Moreover, the time taken for mixing is preferably 1 to 10 minutes or more. Next, the polyether compound is added, and the mixing is further continued to thoroughly mix the components. The mixing may be performed with 25 a conventional mixer. Mixers are classified into batch mixers and continuous mixers. Examples of the batch mixers include open-type rollers, and closed-type banbury mixers, and kneader-type mixers. Examples of the continuous mixers include single-shaft rotor mixers, twin-shaft rotor mixers, 30 single-shaft screw mixers, twin-shaft screw mixers, and multishaft screw mixers. After the mixing, drying and, if necessary, powdering may be performed.

[0041]

35

The lower limit of the ash content in the polyamide resin composition derived from the swelling mica is typically

adjusted to 0.5 percent by weight and preferably 1.0 percent by weight. The upper limit of the ash content is typically adjusted to 50 percent by weight, preferably 40 percent by weight, and more preferably 30 percent by weight. If the ash content is below 0.5 percent by weight, improvement of mechanical properties or warpage reduction may be insufficient, and the upper limit exceeds 50 percent by weight, surface appearance of the resulting product may be deteriorated.

[0042]

5

10

15

20

25

30

35

The structure of the swelling mica dispersed in the polyamide resin composition of the present invention is completely different from the structure of the swelling mica before use. Whereas the swelling mica before the use has micrometer-order aggregated structures consisting of many thin layers, the swelling mica after treatment with the polyether forms independent fine segments as a result of cleaving of the layers. Consequently, the swelling mica becomes dispersed in the polyamide resin composition by forming significantly fine lamellae independent from one another. The number thereof is markedly larger than the number of the swelling mica particles before the use. Such a state of dispersion of the lamellar swelling mica can be expressed by the equivalent circular diameter [D], the aspect ratio (layer length/layer thickness), the number of dispersed particles, the maximum layer thickness, and the average layer thickness.

[0043]

The equivalent circular diameter [D] is defined as the diameter of a circle having the equivalent area as the particle of the swelling mica dispersed in various shapes observed in a micrograph or the like. Of the swelling mica particles dispersed in the polyamide resin composition, those having an equivalent circular diameter [D] of 3000 Å or less is preferably contained in an amount of 20% or more, more preferably 35% or more, yet more preferably 50% or more, and most preferably 65% or more. If the proportion of the swelling mica with the

equivalent circular diameter [D] of 3000 Å or less is lower than 20%, improvement of the mechanical properties of the polyamide resin composition and the warpage reduction may be insufficient. In the polyamide resin composition of the present invention, the average of the equivalent circular diameters [D] of the swelling mica is preferably 5,000 Å or less, more preferably 4,000 Å or less, yet more preferably 4,000 Å or less, and most preferably 3,500 Å or less. If the average of the equivalent circular diameters [D] is larger than 5,000 Å, improvement of the mechanical properties of the polyamide resin composition, warpage reduction may be insufficient, and surface appearance of the resulting product may be deteriorated. The lower limit is not particularly limited. Since no improvement occurs below 100 Å, there is no need to adjust the average equivalent circular diameter to below 100 Å.

[0044]

The equivalent circular diameter [D] may be determined by obtaining a picture of a melt-mixed material, an injection-molded product, or a heat-pressed product using a microscope or the like, arbitrarily selecting from the picture a particular region containing 100 or more of layers of swelling mica, and performing image processing using an image processor or the like so as to allow computer processing for quantitative determination.

[0045]

The average aspect ratio is defined as the number-average of the ratio, layer length/layer thickness, of the swelling mica dispersed in the resin. The lower limit of the average aspect ratio of the swelling mica in the polyamide resin composition of the present invention is preferably 10, more preferably 20, and yet more preferably 30. If the aspect ratio is lower than 10, improving effect of mechanical properties and the like of the polyamide resin composition of the present invention may be insufficient. Since the effect does not change at average aspect ratios exceeding 300, there is no need to increase the

average aspect ratio to beyond 300.

[0046]

5

10

15

25

30

35

Here, the number of the dispersed particles per unit weight ratio of the swelling mica found in an area of 100 µm<sup>2</sup> of the polyamide resin composition is defined as [N] value. [N] value of the swelling mica in the polyamide resin composition of the present invention is preferably 30 or more, more preferably 45 or more, and most preferably 60 or more. upper limit is not particularly limited. However, the effect does not change at [N] values exceeding about 1,000; thus, there is no need to increase the [N] value to over 1,000. For example, [N] values are determined as follows: From a polyamide resin composition, a very thin slice approximately 50 to 100 µm in thickness is cut. A picture of the slice is taken by transmission electron microscopy (TEM) or the like, and the number of particles of the swelling mica found in a desired 100 μm² region in the picture is counted. The number is divided by the weight ratio of the swelling mica used. Alternatively, the [N] value can be determined by selecting a desired region 20 (whose area is measured in advance) containing more than 100 particles from a TEM micrograph, dividing the number of the swelling mica particles in that region by the weight ratio of the swelling mica used, and converting the obtained value to a 100 µm<sup>2</sup> equivalent so as to define this converted value as the [N] value. Thus, the [N] values can be determined by using TEM micrographs or the like of the polyamide resin composition.

[0047]

The average layer thickness is defined as the number-average value of the thickness of the layers of the dispersed lamellar swelling mica. Here, the upper limit of the average thickness of the swelling mica in the polyamide resin composition is preferably 500 Å or less, more preferably, 450 Å or less, and most preferably 400 Å or less. If the average layer thickness is larger than 500 Å, improvement of the mechanical properties and the like of the polyamide resin

composition may be insufficient. The lower limit of the average layer thickness is not particularly limited. However, since no change in effects occurs below  $50\,\text{\AA}$ , there is no need to reduce the average layer thickness to  $50\,\text{\AA}$  or less.

[0048]

5

10

15

20

25

30

35

The maximum layer thickness is defined as the maximum thickness of the layers of the lamellar swelling mica dispersed in the polyamide resin composition of the present invention. Here, the upper limit of the maximum layer thickness is preferably 2,000 Å or less, more preferably 1,800 Å or less, and most preferably 1,500 Å or less. If the maximum layer thickness is larger than 2,000 Å, mechanical properties of the polyamide resin composition and surface appearance of the resulting product may be deteriorated. The lower limit of the maximum layer thickness of the swelling mica is not particularly limited but is preferably at least 100 Å, more preferably at least 150 Å, and most preferably at least 200 Å.

[0049]

The layer thickness and the layer length can be determined from a micrograph or the like of a film prepared by heat-press-molding or draw-molding a polyamide resin composition of the present invention melted by heating or from a picture of a thin product prepared by injection-molding using a molten resin. In particular, assume that a film prepared as above or an injection-molded thin, flat specimen having a thickness of approximately 0.5 to 2 mm is placed on the X-Y plane. From this film or the specimen, a very thin slice having a thickness of approximately 50 to 100 µm is cut out along a plane parallel to the X-Z plane or the Y-Z plane, and this slice is observed at high magnifications of about 40,000 to 100,000 or more by transmission electron microscopy or the like to determine these values. Alternatively, these values can be determined by arbitrarily selecting a region containing 100 or more particles of the swelling mica from a TEM micrograph taken as in the above and performing the image processing to allow

computer processing for quantitative determination. Alternatively, a ruler or the like may be used for the determination.

[0050]

5

10

15

20

25

30

35

The method for producing the polyamide resin composition of the present invention is not particularly limited. For example, the polyamide resin composition may be made by melt-mixing the polyamide resin and the swelling mica treated by the polyether compound using various types of conventional mixers. Examples of the mixer include single-shaft extruders, twin-shaft extruders, rollers, banbury mixers, and kneaders. Mixers having high shear efficiency are particularly preferable. The polyamide resin and the swelling mica treated with the polyether compound may be simultaneously placed in the above-described mixer. Alternatively, the polyamide resin may be melted in advance and then melt-mixed with the swelling mica.

[0051]

If necessary, the polyamide resin composition of the present invention may contain polybutadiene, a butadiene-styrene copolymer, acryl rubber, ionomer, an ethylene-propylene copolymer, an ethylene-propylene-diene copolymer, natural rubber, chlorinated butyl rubber, an  $\alpha$ -olefin homopolymer, a copolymer of two or more  $\alpha$ -olefins (the copolymer may be a random copolymer, a block copolymer, a graft copolymer, or the like, or may be a mixture of these), or an impact resistance improver such as an olefin elastomer. may be modified by an acid compound such as maleic anhydride or an epoxy compound such as glycidyl methacrylate. As long as the mechanical properties and the like are not adversely affected, any other thermoplastic resin or thermosetting resin may be used. Examples thereof include unsaturated polyester resins, polyester carbonate resins, liquid crystalline polyester resins, polyolefin resins, thermoplastic polyester resins, rubber polymer-reinforced styrene resins, polyphenylene sulfide resins, polyphenylene ether resins,

polyacetal resins, polysulfone resins, and polyarylate resins. These may be used alone or in combination.

[0052]

5

20

25

Depending on the purpose, various additives, such as pigments and dyes, a heat stabilizer, an antioxidant, a UV absorber, a photostabilizer, a lubricant, a plasticizer, a flame retarder, and an antistatic agent, may be added.

[0053]

The polyamide resin composition of the present invention is suitable for injection molding and heat-press molding and can be used in blow molding. The resulting product has excellent appearance, satisfactory mechanical properties, and high resistance to thermal deformation. Thus, for example, the composition is suitable for use in automobile parts, parts for home appliances, domestic housewares, wrapping materials, and other general industrial materials.

[0054]

[EXAMPLES]

The present invention will now be described in detail with reference to the examples below; however, the present invention is not limited to these examples.

[0055]

Below is a summary list of the primary materials used in EXAMPLES and COMPARATIVE EXAMPLES. Note that these materials were not purified unless otherwise noted.

(Materials)

- Polyamide resin A (Nylon 6): Unitika Nylon 6 A1030BRL (product of Unitika Ltd.)
- Polyamide resin B (Nylon 66): Unitika Nylon 66 A125N (product of Unitika Ltd.)
  - Polyamide resin C (Nylon 46): Stanyl TS300 (product of DSM-JSR)
- Polyamide resin D (Nylon MXD6): Reny 6002 (product of Asahi 35 Kasei Corporation)

- Swelling mica E: Somasif ME100 (product of Co-op Chemical Co., Ltd.)
- Polyether compound G: BISOL 18EN (product of Toho Chemical Industry Co., Ltd.)
- 5 - Polyether compound H: BISOL 20PN (product of Toho Chemical Industry Co., Ltd.)

[0056]

10

15

20

25

30

(Determining Dispersion State)

Dispersion state was determined using an ultrathin section 50 to 100 µm in thickness obtained by a frozen section technique. The section was observed with a transmission electron microscope (JEM-1200EX, product of JEOL) with an accelerating voltage of 80 kV at a magnification of 40,000 to 1,000,000 to obtain a microgram of the dispersion state of swelling mica. From this TEM micrograph, a region containing 100 or more of dispersed particles was arbitrarily selected. The layer thickness, the layer length, and the number of particles ([N] values) were determined either manually with a graduated ruler or by processing the microgram with Image Analyzer PIAS III produced by Inter Quest Co., Ltd.

The equivalent circular diameter [D] was determined with Image Analyzer PIAS III produced by Inter Quest Co., Ltd. [0057]

The [N] values were determined as follows. First, the number of particles of the swelling mica present in the selected region in the TEM micrograph was determined. The ash content of the resin composition derived from the swelling mica was separately determined. The number of the particles was divided by the ash content and the resultant value of number per 100 um<sup>2</sup> area was determined as the [N] value. The number-average value of the layer thickness of the swelling mica was assumed as the average layer thickness; and the maximum value among the layer thickness of the swelling mica was assumed as the maximum layer thickness. A sample containing large dispersed particles and thus not suitable for observation by TEM was

35

examined using an optical microscope (Optical microscope BH-2 produced by Olympus Optical Co., Ltd.), and the [N] values were determined as is described above. Where required, a sample was melted at 250°C to 270°C using Hotstage THM 600 produced by Linkam Scientific Instruments Ltd., and the state of the dispersed particle was observed in a molten state. The number-average value of the ratios of layer length to layer thickness of the individual swelling mica particles was assumed as the average aspect ratio. For dispersed particles that were not lamellar, the aspect ratio was determined as the ratio, major axis/minor axis. Here, a "major axis" is the long side of a hypothetic rectangle having the minimum area among hypothetic rectangles circumscribed with a target particle in a micrograph, and a "minor axis" is the short side of this hypothetic rectangle.

[0058]

10

15

20

25

30

(Flexural Properties)

The polyamide resin composition of the present invention was dried (90°C, 10 hours). Using an injection molding machine operating at a clamping pressure of 75 tons, a specimen having dimensions of approximately 10 x 100 x 6 mm was formed by injection molding at a resin temperature in the range of 240°C to 300°C. (Nylon 6: 240°C, Nylon 66: 260°C, MXD6: 280°C, Nylon 46: 300°C). The flexural strength and the flexural elastic modulus of the specimen were measured according to ASTM D-790.

[0059]

(Deflection Temperature Under Load)

The deflection temperature under load was determined according to ASTM D-648 under a load of 1.86 MPa by using the same type of specimen used in determining the flexural properties.

[0060]

(Warpage)

The polyamide resin composition of the present invention was dried (90°C, 10 hours). A tabular specimen having

dimensions of approximately 120 x 120 x 1 mm was formed by injection molding at a resin temperature in the range of 240°C to 300°C. (Nylon 6: 240°C, Nylon 66: 260°C, Nylon MXD6: 280°C, Nylon 46: 300°C). The tabular specimen was placed on a flat surface, and one of the four corners was held down. Of the remaining three corners, one most distant from the flat surface was determined by measuring the distance with a slide caliper. The same was repeated for every corner, and the obtained warpages were averaged.

10 [0061]

5

(Mold Shrinkage Rate)

The polyamide resin composition of the present invention was dried (90°C, 10 hours). A tabular specimen having dimensions of approximately 120 x 120 x 2 mm was formed by injection molding at a resin temperature in the range of 240°C to 300°C. (Nylon 6: 200°C, Nylon 66: 260°C, MXD6: 280°C, Nylon 46: 300°C). The mold shrinkage rate was determined by the following equation. In the table below, MD denotes the direction of the resin flow, and TD denotes a direction orthogonal to the resin flow.

Mold shrinkage rate (%)

=(size of die-observed size of molded product)/(size of die)  $\times$  100

25

15

20

(Centerline Average Roughness)

The centerline average roughness was determined with surface analyzer Surfcom 1500A produced by Tokyo Seimitsu Co., Ltd., using the same type of above specimen.

30 [0062]

(Ash Content)

The ash content of the polyamide resin composition derived from the swelling mica was determined according to JIS K 7052.

35 [0063]

(Method B flow)

The polyamide resin composition of the present invention was dried (90°C, 10 hours). The method B flows after 5 minutes and after 15 minutes were determined using a flow tester produced by Shimadzu Corporation at a temperature of 280°C. under a load of 100 kg. The melt stability is higher when the difference between the flow after 5 minutes and the flow after 15 minutes is smaller.

[0064]

### 10 (SYNTHETIC EXAMPLE 1)

Ion-exchange water, polyether compounds, and swelling mica at the weight ratios set forth in Table 1 were mixed for 15 to 30 minutes. The mixture was then dried and powdered to prepare swelling mica (J-1 to J-4) treated with the polyether compounds.

[0065]

[Table 1]

15

20

25

30

	Clay J-1	Clay J-2	Clay J-3	Clay J-4
Water	100	100	100	100
Swelling mica E	. 8	8	8	8
Polyether compound G	4	1.6	0.8	
Polyether compound H		-		2.5

(unit: parts by weight)

[0066]

# (SYNTHETIC EXAMPLE 2)

Ion-exchange water, swelling mica, polyvinylpyrrolidone (PVP), methylstearylbis[PEG]ammonium chloride (Ethoquad produced by Lion Akzo Co., Ltd.), and trioctylmethylammonium chloride at the weight ratios set forth in Table 2 were mixed for 15 to 30 minutes. Subsequently, the mixture was dried and powdered (K-1 to K-3).

35 [0067]

[Table 2]

	Clay K-1	Clay K-2	Clay K-3
Water	100	100	100
Swelling mica E	8	8	8
Polyvinylpyrrolidone	4		
Methylstearylbis[ PEG] ammonium chloride		8	
Trioctylmethylammonium chloride			4

(unit: parts by weight)

10

15

20

25

30

5

[0068]

(EXAMPLES 1 and 2 and COMPARATIVE EXAMPLES 1 to 4)

Polyamide A, the swelling mica prepared in SYNTHETIC EXAMPLE 1 (J-2 and J-4), the swelling mica E and the swelling mica prepared in SYNTHETIC EXAMPLE 2 (K-1 to K-3) at the weight ratios set forth in Table 3 were melt-mixed using a twin shaft extruder (TEX 44 produced by Japan Steel Works, Ltd.) while adjusting the temperature to the dies to 220°C to 250°C from the initial stage of the mixing. The physical properties of the resulting polyamide resin compositions were evaluated. The results are shown in Table 3.

Table 3 shows that the swelling mica treated with PVP or ammonium salts did not exhibit sufficient reinforcing effect. Moreover, the warpage and the mold shrinkage were not significantly improved, and deterioration at the processing temperature was significant.

[0069]

# (COMPARATIVE EXAMPLE 5)

To a pressure reactor, 5,000 g of  $\epsilon$ -caprolactam, 1,100 g of water, and 550 g of swelling mica were placed, and heated to 250°C while stirring. The pressure inside the reactor was increased to 4 kg/cm² to 15 kg/cm² while releasing steam. The pressure was then reduced to about 2 kg/cm² and the temperature was increased to about 260°C. Under these conditions,

35 polymerization for a polyamide composition containing

approximately 10% of swelling mica was attempted. In the midst of the reaction, the motor load current of the mixer of the reactor became unstable, which eventually led to mixing failure due to overload. Thus, the polymerization was discontinued. In other words, in a polymerization method, an attempt was made to obtain a polyamide composition by polymerization but failed due to an increased melt viscosity.

[0070]

5

[Table 3]

											_	_						_					_	$\neg$			7	
5		5							11	10.1	144	i	i	Ī	:	•	248	***	844	Ŧ		;	***	***		ŧ	Į	Polymerization
	PLES	4						18		10.2	2	13400	ထ	3(*1)	App. 3,000 (*2)	App. 10,000 (*3)	3200	80	79	7.8		1.42	1.59	200		78	:	Melt mixing
10	COMPARATIVE EXAMPLES	3					52			10.0	15	2200	. 25	æ	580	2250	3430	42	82	5.9		\$	1.45	98.0		112	ı	Melt mixing
15	NOO	2	5			18				10.2	5	12300	10	7	890	4500	-	•	•	8		•	•	•		:	••	Melt mixing
		1							÷	10.0	0	24200	က	1.5 (*1)	App. 20,000 (*2)	App. 700,000 (*3)	3010	92	20	7.8		1.43	1.59	210	-	98	35	Melt mixing
20	EXAMPLES	2			15					9.6	81	1170	113	38	95	380	5100	130	139	1.6		0.47	0.53	4.0		32	31	Mell mixing
	EXAIN	-		13												330			ŀ	ı			0.49	4.0		31	30	Melt mixing
25			Parts by weight	•						%;м	%	.4	no./wt% ·100 µm²	۔ ا	.A		MPa	MPa	္စ	mm	%			υu	×10-2 ml/sec			
30					i						Ratio of [D] ≤ 3,000 Å		spersed particles				Flexural elastic modulus		ature under lo			QW	σz	Centerline average roughness	M(	After 5 mln	After 15 min	
35			Polyamide A	Clay J-2	Clay J-4	Clay K-1	Clay K-2	Clay K-3	Swelling mica E	Ash content	Ratio of [D]	Average (D)	Number of a	Average aspect ratio	Average layer thickness	Maximum la	Flexural elas	Flexural strength	Deflection le	Warpage	Mold shrinkage rate			Centerline a	Method B flow			Note

- \*: Significant deterioration occurred during the melt processing; thus, no specimen usable in measurement was obtained.
- \*\*: Measurement was not possible due to severe deterioration.
- \*\*\*: The mixer motor of the reactor was overloaded due to an increase in the melt viscosity, and the polymerization was thus discontinued.
  - (\*1): Because dispersed particles were not lamellar, the ratio, major axis/minor axis, of the dispersed particles was determined.
  - (\*2): Because dispersed particles were not lamellar, the number-average value of the minor axes of the dispersed particles was determined.
- (\*3): Because dispersed particles were not lamellar, the
  maximum value of the minor axis of the dispersed particles was
  determined.

[0071]

### (COMPARATIVE EXAMPLES 6 to 8)

Polyamide A, talc, mica, and a glass fiber reinforcing material at the weight ratios set forth in Table 4 were melt-mixed as in EXAMPLE 1. Evaluation of the resulting polyamide resin compositions was carried out. The properties thereof were evaluated. The results are shown in Table 4.

[0072]

10

and the surface quality was degraded. Although the reinforcing effect could be achieved by adding the glass fibers, this adversely affected the warpage and the surface quality. Thus, none of COMPARATIVE EXAMPLES 5 to 7 achieved a satisfactory balance.

[0074]

(EXAMPLES 3 to 6)

Polyamide A and the swelling mica obtained in SYNTHETIC EXAMPLE 1 at the weight ratios set forth in Table 5 were melt-mixed using a twin shaft extruder (TEX 44 produced by Japan Steel Works, Ltd.) to obtain polyamide resin compositions. Evaluation of the resulting polyamide resin compositions was carried out. The results are shown in Table 5.

[0075]

15 [Table 5]

5

10

			EXAMPLES				
			3	4	5	6	
	Polyamide A	Parts by		10	00		
	Clay J-2	weight	5	7.8	20	30	
	Ash content	wt%	4.0	6.0	13.9	19.2	
20	Ratio of [ D] ≤ 3,000 Å	8	90	90	80	65	
20	Average [ D]	À	1030	1040	1400	1680	
	Number of dispersed	no./wt%	135	135	112	85	
	particles [N]	∙100 μm²					
	Average aspect ratio	-	138	140	102	82	
	Average layer	À	83	85	110	130	
	thickness	,					
	Maximum layer	Å	340	330	480	650	
25	thickness						
	Flexural elastic	MPa	4010	4850	6650	8030	
	modulus						
	Flexural strength	MPa	118	123	130	132	
	Deflection temperature	°C .	108	131	160	182	
	under load						
	Warpage	mm	2.5	1.9	1.0	0.6	
30	Mold shrinkage rate	g <sub>o</sub>					
	MD		0.67	0.54	0.29	0.19	
	TD		0.68	0.59	0.32	0.20	
	Centerline average	nm	3.8	3.8	4.3	5.7	
	roughness						

[0076]

35 (EXAMPLES 7 to 11)

Polyamide B and the swelling mica obtained in SYNTHETIC EXAMPLE 1 at the weight ratios set forth in Table 6 were melt-mixed using a twin shaft extruder (TEX 44 produced by Japan Steel Works, Ltd.) while adjusting the temperature to the dies to 230°C to 260°C from the initial stage of the mixing. Evaluation of the resulting polyamide resin compositions was carried out. The results are shown in Table 6.

[0077]

[Table 6]

5

	L	1	EXAMPL	ES		
		7	8	9	10	11
Polyamide B	Parts by			100		
Clay J-1	weight	16				
Clay J-2			13	28	1	i
Clay J-3					11	
Clay J-4					l	14
Ash content	wt8	9.3	9.5	18.1	9.1	9.4
Ratio of [D] $\leq 3,000$		92	80	70	69	35
Average [ D]	Å	1080	1450	1650	2010	4170
Number of dispersed	no./wt%	140	100	92	80	46
particles [N]	-100 μm²					
Average aspect ratio	-	143	105	95	88	51
Average layer thickne		78	105	125	145	345
Maximum layer thickne	Š Å	300	500	580	650	1520
Flexural elastic	MPa	5900	5600	7990	5550	4850
modulus						
Flexural strength	MPa	125	115	131	115	91
Deflection temperature	e °C	148	140	178	138	123
under load						
Warpage	mm	1.0	1.0	0.5	1.2	3.7
Mold shrinkage rate	8	1				
	4D	0.25	0.30		0.36	0.67
	rD	0.27	0.33	0.17	0.40	0.70
Centerline average	nm	3.7	4.1	5.2	4.0	7.0
roughness				ļ		

[0078]

# (COMPARATIVE EXAMPLES 9 to 11)

Polyamide B, talc, mica, and a glass fiber reinforcing material at the weight ratios set forth in Table 7 were melt-mixed as in EXAMPLE 1. Evaluation of the resulting polyamide resin compositions was carried out. The results are shown in Table 7.

35 [0079]

[Table 7]

			COMPARATIVE EXAMPLES					
			9	10	11			
	Polyamide B	Parts by		100				
	Talc	weight	11					
5	Mica			11				
	Glass fibers				11			
	Ash content	wt%	10.0	10.0	10.0			
	Ratio of [ D] ≤ 3,000	ક્ર	0	0	Not			
	À				measured			
	Average [ D]	Â	24200	25800	Not			
					measured			
1.0	Number of	no./wt%	5	2	Not			
10	dispersed	·100 µm²			measured			
	particles [N]							
	Average aspect	_	1.5 (*1)	1.5 (*1)	Not			
	ratio	•			measured			
	Average layer	Å	App. 30,000	App. 30,000	Not			
	thickness	À	(*2)	(* 2)	measured			
	Maximum layer	A	App.	App. 900,000	Not			
15	thickness		900,000	(* 3)	measured			
10			(* 3)	4500	5500			
	Flexural elastic	MPa	3200	4500	5500			
	modulus Flexural strength	MPa	108	123	135			
	Deflection	°C	86	97	148			
	temperature under	C	00	91	140			
	load							
0.0	Warpage	mm	7.2	6.5	13.5			
20	Mold shrinkage	8						
	rate			•				
	MD		1.38	1.30	0.44			
	TD		1.49	1.41	1.56			
	Centerline average	nm	200	430	700			
	roughness							

- 25 (\*1): Because dispersed particles were not lamellar, the ratio, major axis/minor axis, of the dispersed particles was determined.
  - (\*2): Because dispersed particles were not lamellar, the number-average value of the minor axes of the dispersed particles was determined.
  - (\*3): Because dispersed particles were not lamellar, the maximum value of the minor axis of the dispersed particles was determined.

[0080]

30

35

Table 7 shows that neither talc nor mica conventionally

used exhibited a sufficient reinforcing effect. Moreover, the warpage and the mold shrinkage were not significantly improved, and the surface quality was degraded. Although the reinforcing effect could be achieved by adding the glass fibers, this adversely affected the warpage and the surface quality. Thus, none of COMPARATIVE EXAMPLES 8 to 10 achieved a satisfactory balance.

[0081]

10

15

(EXAMPLE 12 and COMPARATIVE EXAMPLE 12)

Polyamide C and the swelling mica prepared in SYNTHETIC EXAMPLE 1 or talc at the weight ratios set forth in Table 8 were melt-mixed using a twin shaft extruder (TEX 44 produced by Japan Steel Works, Ltd.) while adjusting the temperature to the die to 280°C to 300°C from the initial stage of mixing. Evaluation of the resulting polyamide resin compositions was carried out. The results are shown in Table 8.

[0082]

[Table 8]

			EXAMPLE	COMPARATIVE
				EXAMPLE
		<del></del>	12	12
	Polyamide C	Parts by		100
5	Clay J-2	weight	13	
	Talc			11
	Ash content	wt&	9.6	10.0
	Ratio of [ D] ≤ 3,000 Å	¥	89	0
	Average [ D]	Å .	1025	24200
	Number of dispersed	no./wt%	134	5
10	particles [N]	·100 µm²		
10	Average aspect ratio	_	140	1.5 (*1)
	Average layer	Å	85	App.30,000
	thickness	2	ļ	(* 2)
	Maximum layer	Å	340	App. 900,000
	thickness			(* 3)
	Flexural elastic	MPa	5080	5150
15	modulus			
	Flexural strength	MPa	145	135
	Deflection	. °C	186	135
	temperature under			
	load		0.0	1 0
	Warpage	mm %	0.8	1.9
	Mold shrinkage rate   MD	8	0.35	0.54
20	TD		0.45	0.59
	Centerline average	nm	3.8	210
	roughness	11111	3.0	210
		L	l	L

- (\*1): Because dispersed particles were not lamellar, the ratio,
  25 major axis/minor axis, of the dispersed particles was
  determined.
  - (\*2): Because dispersed particles were not lamellar, the number-average value of the minor axes of the dispersed particles was determined.
- 30 (\*3): Because dispersed particles were not lamellar, the maximum value of the minor axis of the dispersed particles was determined.

[0083]

Table 8 shows that the talc conventionally used did not exhibit a sufficient reinforcing effect. Moreover, the

warpage and the mold shrinkage were not significantly improved, and the surface quality was degraded.

[0084]

# (EXAMPLE 13 and COMPARATIVE EXAMPLE 13)

Polyamide D and the swelling mica prepared in SYNTHETIC EXAMPLE 1 or talc at the weight ratios set forth in Table 9 were melt-mixed using a twin shaft extruder (TEX 44 produced by Japan Steel Works, Ltd.) while adjusting the temperature to the dies to 250°C to 290°C from the initial stage of the mixing.

Evaluation of the resulting polyamide resin compositions was carried out. The results are shown in Table 9.

[0085]

[Table 9]

5

10

		EXAMPLE	COMPARATIVE EXAMPLE
		13	13
Polyamide D	Parts by		100
Clay J-2	weight	13	
Talc			11
Ash content	wt%	9.5	10.0
Ratio of [D] $\leq 3,000  \text{Å}$	o <sub>o</sub>	102	0
Average [ D]	Ä	1320	24200
Number of dispersed	no./wt%	120	5
particles [N]	·100 μm²		
Average aspect ratio	_ `	125	1.5 (*1)
Average layer	Å	100	App. 30,000
thickness			(*2)
Maximum layer	Å	410	App. 900,000
thickness			(* 3)
Flexural elastic	MPa	7000	4910
modulus			
Flexural strength	MPa	167	135
Deflection	°C	116	88
temperature under			
load			1.0
Warpage	mm	1.0	1.9
Mold shrinkage rate	ક	0.45	3.46
MD		0.45	1.46
TD		0.54	1.53
Centerline average roughness	nm	4.1	220
Loughtess	L	L	L

35

- (\*1): Because dispersed particles were not lamellar, the ratio, major axis/minor axis, of the dispersed particles was determined.
- (\*2): Because dispersed particles were not lamellar, the number-average value of the minor axes of the dispersed particles was determined.
  - (\*3): Because dispersed particles were not lamellar, the maximum value of the minor axis of the dispersed particles was determined.

10 [0086]

15

20

Table 9 shows that the talc conventionally used did not exhibit a sufficient reinforcing effect. Moreover, the warpage and the mold shrinkage were not significantly improved, and the surface quality was degraded.

[0087]

[EFFECT OF THE INVENTION]

As is described above, the swelling mica treated with the polyether compound according to the present invention achieves homogeneous, fine dispersion in the polyamide resin. Thus, a polyamide resin composition exhibiting low warpage, good surface appearance, improved mechanical properties, and high heat resistance can be obtained while achieving a satisfactory balance between these properties.

[Document Name] Abstract
[Abstract]

[Problem to be solved by the Invention] To provide a polyamide resin composition which is inhibited from warpage after molding, gives a molded article having a satisfactory surface appearance, high mechanical properties, and high heat resistance, and has an excellent balance among material properties; and a process for producing the polyamide resin composition.

[Means for solving problem] The polyamide resin composition is obtained by melt-kneading a polyamide resin and a swellable mica treated with a polyether compound having a bisphenol structure. The process for producing a polyamide resin composition is characterized by melt-kneading a polyamide resin together with a polyether compound.

[Selective Figure] none